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Measuring of Q-values for (αp)-reactions on aluminium and boron by means of photographic emulsions

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With 14 figures in the text

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1. Introduction. The (αp)-reaction is, as is well known, the first observed artificial disintegration process (1). An α-particle is captured by a nucleus and a proton is emitted. The reaction is of the form (2)

$$Z^{A} + \alpha \rightarrow (Z + 1)^{A+3} + p + Q. \tag{1}$$

The quantity Q represents the mass defect. Q is > 0 if energy is released by the reaction, but < 0 if energy is absorbed. Q is the excess of the sum of the kinetic energy of the proton and that of the recidual nucleus over the energy of the incident α-particle. Z and A are the atomic number and the mass number, respectively.

If M_α, M_p and M_r and E_α, E_p and E_r represent the masses and energies, respectively, of the α-particle, the proton and the residual nucleus, we obtain from the usual momentum and energy conservation equations (2, 3)

$$Q = E_p\left(1 + \frac{M_p}{M_r}\right) - E_\alpha\left(1 - \frac{M_\alpha}{M_r}\right) - \frac{2\sqrt{E_p\, E_\alpha\, M_p\, M_\alpha}}{M_r}\cos\vartheta \tag{2}$$

where ϑ is the angle between the direction of motion of the proton and that of the α-particle.

By measuring the ranges of the protons and the angles ϑ it is possible to compute Q, when the masses of the nuclei in the reaction and the energy of the α-particle are known. In doing this we may obtain several different Q-values corresponding to different excited states in which the residual nucleus may find itself. These excited states decay to the ground state by emission of one or more γ-quanta. The ground state corresponds to the highest Q-value which, however, may not be observed in the experiment performed.

α-particles, the energy of which is insufficient to surmount the Gamow wall, are frequently able at certain energies to penetrate into the nucleus and produce disintegration (resonance disintegration).

The protons are usually for the sake of simplicity observed "at right angle" ($\vartheta = 90^\circ$) or "in forward direction" ($\vartheta = 0^\circ$).

If we know the mass of one of the nuclei with atomic number either Z or $Z + 1$, and if we have measured the Q-value, corresponding to the ground state, it is, according to equation (1), possible to calculate the mass of the other nucleus.

The energy of the proton and that of the α-particle can be measured by magnetic deflection or by the range in air, some gas or a photographic emulsion. If the protons are observed for a certain value ϑ and the energy of the α-particles is constant, the ranges of the protons are grouped, each group belonging to a certain Q-value. The ranges increase when the α-particle energy increases, and new proton groups corresponding to lower Q-values may appear while others disappear. It will sometimes be of interest to know if γ-radiation accompanies the disintegration, and, if this is the case, how the intensity of the radiation varies as a function of the energy of the incident α-particles or at least at what energy the γ-radiation begins (4).

It will be seen from the next section that many $(\alpha\, p)$-reactions have been investigated by several workers. Proton groups are observed at various α-particle energies. The results of different authors are often inconsistent. This is especially the case in the first experiments with the natural α-particle sources, partly as a consequence of the spread of the energy of the α-particles, partly as a consequence of the "poor geometry".

The photographic emulsions have up to now not been used in connection with $(\alpha\, p)$-reactions only except for recording the number of protons and measuring the energy of these protons. The purpose of this work is to investigate the possibility of using the emulsions also for determining the paths of the individual protons and (indirectly) the paths of the corresponding α-particles and in this way to improve the geometry. (In other words, we have tried to replace the Wilson chamber by the photographic emulsions for the study of the $(\alpha\, p)$-reactions.)

2. Earlier experiments. In the present section those works will mainly be quoted which are of special interest in connection with the $(\alpha\, p)$-reaction on aluminium and boron as well as methods for the study of $(\alpha\, p)$-reactions by means of natural α-particles.

In 1919 RUTHERFORD (1, 5) bombarded light elements with α-particles from RaC' with the view of investigating whether some lighter atoms could be disintegrated by the intense forces brought into play in such close collisions. As is well known the first reaction was observed on nitrogen.

RUTHERFORD and CHADWICK (6) found furthermore that in addition to nitrogen also boron, fluorine, sodium, aluminium and phosphorus exposed to α-rays gave rise to protons whose ranges vary between 40 and 90 cm of air. In these experiments the material subjected to the bombardment was placed immediately in front of the source of α-particles, and microscopical observations of the ejected particles were made on a zinc sulphide screen placed in a direct line a few centimetres away. The ranges of the H-particles were greater than the range (30 cm in air) of free H-nuclei set in motion by α-particles so that, by inserting absorbing screens of 30 cm air equivalent, the results were made independent of the presence of hydrogen as an impurity in the bombarded material.

To overcome the difficulties of investigating low energy disintegration protons in the presence of recoil protons from hydrogen in the target material or window, or in the presence of high energy α-particles in the source, RUTHERFORD and CHADWICK (8) devised a simple method for observing ejected protons having a range of only 7 cm in air. This method consisted in examining the liberated particles at an average angle of 90° to the direction of the incident α-particles. By means of screens it was arranged that no α-particles could fall directly on the zinc sulphide screen. RUTHERFORD and CHADWICK found that in addition to the abovementioned elements a number of other elements gave particles of ranges above 7 cm, namely: neon, magnesium, silicon, sulphur, chlorine, argon and potassium.

In 1920 RUTHERFORD (5) suggested the possibility to obtain further information about the nuclei by a study of the tracks of α-particles through oxygen or nitrogen by the well-known expansion method of WILSON. BLACKETT (9) obtained evidence by Wilson photographs that in the case of nitrogen the α-particle is captured when a proton is expelled. The number of photographs needed is very large. Blackett studied 23 000 photographs with an average number of 18 α-tracks (range 8.6 cm) on each film. Two photographs at right angles were taken by each exposure. Amongst the normal forks due to elastic collisions, only 8 were found of the (αp)-reaction type. Blacket's experiment showed, that an α-particle that ejects a proton from a nitrogen nucleus is itself bound to that nucleus.

SCHMUTZER (10) used the Geiger point counter for recording the protons from aluminium bombarded with RaC′ α-particles. The experimental equipment was suggested by RAUSCH v. TRAUBENBERG. The range of the protons (89 cm) was in agreement with that (90 cm) obtained by Rutherford.

BOTHE and FRÄNZ (11) also used a point counter. The elements from boron to calcium (except neon and argon) were bombarded with α-particles from polonium (range 3.8 cm). Disintegrations were obtained for boron, nitrogen, magnesium and aluminium. The proton ranges in air for nitrogen and aluminium were 16 cm. Later BOTHE and FRÄNZ (12) found two proton groups for boron. The corresponding ranges were 20 and 32 cm respectively. The reason for those groups was assumed to be the occurrence of the two well-known boron isotopes.

The yield of protons from aluminium of 0.5 cm air equivalent bombarded with α-particles of 7 cm range is one in a million according to Rutherford and Chadwick and about the same according to Bothe and Fränz.

FRÄNZ (13) bombarded boron with α-particles from RaC' as well as from polonium and studied the protons emitted in the backward direction. In both cases two proton groups were found, the longer of which represented a third proton group ejected from boron. BOTHE (14) confirmed the existence of these three groups. The yield of the longer groups, 33 and 74 cm, were 6.5×10^{-6} and 0.17×10^{-6} respectively. As a possible explanation of the different proton groups Bothe proposed among other things the emission of a γ-quantum. BECKER and BOTHE (15) computed the energy of the γ-radiation from B from measurements of the absorption in Al of the β-particles released by the γ-radiation. They found the value 3.1 MeV. This agrees with the difference between the Q-values for the disintegrations corresponding to the two longer proton groups. Furthermore, the γ-radiation appears at the same α-particle energy at which the lower proton group starts.

RUTHERFORD and CHADWICK (16) studied absorption curves for protons from Al ($\vartheta = 90°$) bombarded with α-particles from RaC, and found Q-values which differed by up to 5 or 6 MeV. They concluded that either the mass of the aluminium nucleus or that of the newly formed silicon nucleus or both were not constant.

POSE (17), using an ionization chamber in connection with a Hoffmann electrometer, bombarded a thick aluminium layer with Po-α ($\vartheta = 0°$). Protons of three different ranges occurred (30 cm, 47 cm, > 60 cm for α-ranges of 2.50, 2.93 and 3.72 cm respectively). The shortest group (18) occurred at all α-particle energies and the range of the group increased as the energy of the α-particle increased. The longer proton groups were pure resonance groups, which appeared only for definite α-particle energies.

CHADWICK, CONSTABLE and POLLARD (19) adopted an ionization chamber in connection with an amplifier and an Einthoven string galvanometer for the detecting of the protons. They bombarded Al, B, C, N, O, F, Na, Mg, Al, Si, P and S with α-particles from polonium. The emission of protons was established from B, N, F, Na, Al and P. The protons were observed both in forward direction and at right angle. The Q-value, that is the release of energy, was now introduced. For boron the two proton groups observed gave the values 3.2 MeV and 0.2 MeV respectively. For the third proton group found by Bothe and Fränz, they computed $Q = -1.4$ MeV. The isotope B^{10} was assumed to be responsible for the proton groups.

Chadwick, Constable and Pollard found for the proton groups from aluminium $Q = 2.3$ MeV and -0.1 MeV respectively.

CHADWICK and CONSTABLE (20) reexamined the investigations on aluminium and fluorine using better geometry, which was possible by introducing a stronger polonium source. They resolved the proton emission from aluminium into eight distinct groups and showed that these groups were due to penetration of the α-particle through four resonance levels, each giving rise to a pair of groups. The Q-value for the shorter groups was about 0 to 0.1 MeV, while that for the longer group was about 2.3 MeV.

DIEBNER and POSE (21) repeated and confirmed the earlier measurements by Pose. The sharp proton groups from aluminium did not confirm the results obtained by Chadwick and Constable. Both resonance groups gave the Q-value 3.53 MeV, the lowest group gave an average of 0.03 MeV.

STEUDEL (22) confirmed the three proton groups from aluminium bombarded with polonium α-particles. He also found traces of a fourth group. However, he could not confirm the resonance character of the longer groups.

HEIDENREICH (23) used an ionization chamber with an electrometer, almost the same arrangement as Pose, and bombarded boron with polonium α-particles. The protons were observed in the forward direction. He found three proton groups, the Q-values of which were -1.92, 1.27 and 7.63 MeV respectively. The second value is, however, an average of four Q-values between 0.76 and 1.6, obtained for four different energies of the α-particles. The yield for the second group is 4×10^{-6} and that for the third group 0.2×10^{-6}. The air equivalent of the boron target was 12 mm.

PATON (24) bombarded boron with α-particles from ThC′ and observed the protons at right angle with a Geiger proportional counter. On the assumption that the α-particles are captured by B^{10} the Q-values obtained were 3.1, 0.35, -0.78 and -1.86 MeV.

von BAEYER (25) showed by coincidence measurements that the γ-radiation from B, bombarded with Po-α, was associated mainly or entirely with the 33 cm or second proton ground. Thus the explanation of Becker and Bothe (l. c.) was confirmed.

HAXEL (26) bombarded aluminium with α-particles from RaC and ThC. The protons emitted were detected by means of a proportional counter. The number of protons was studied as a function of the α-particle range (4.6—8.6 cm). He found two groups, the Q-values of which were about 0 and 2.3 MeV. For α-particles of 8.6 cm range there appeared a third proton group, with a Q-value of about -2 MeV. Later HAXEL (27, 28) found four proton groups, with $Q = 2.3$, 0, -1.1 and -2.6 MeV, respectively.

HAFSTAD (29) confirmed the resonance character of the longer proton groups from aluminium found by Pose. He used a polonium α-source, an ionization chamber and an improved electric circuit of Du Bridge-type.

DUNCANSON and MILLER (30) used a Geiger-Klemperer ball counter to detect the protons from aluminium and from magnesium bombarded with α-particles from RaC′. They found for aluminium four proton groups, corresponding to the Q-values 2.07, -0.16, -1.53 and -2.67 MeV respectively. LIVINGSTON and BETHE (l. c.) have corrected these values for the thickness of the sample and got 2.26, -0.02, -1.32 and -2.49 MeV, respectively.

MILLER, DUNCANSON and MAY (31) found for boron the Q-values 3.1, 0.4, -0.1 and -1.0 MeV. According to Livingstone and Bethe (l. c.) these values should be corrected for the thickness of the boron layer and replaced by 3.3, 0.5, 0.1 and -0.8.

LIVINGSTON and BETHE (l. c.) called attention to a discrepancy between the highest observed Q-value 3.1 MeV and the expected energy of disintegration into the ground state of the C^{13} residual nucleus calculated from known masses, which is 4.15 MeV. This discrepancy suggests that the group representing the transition into the ground state C^{13} had not been observed. BRUBAKER and POLLARD (32) searched for such a group and found one of very weak intensity, corresponding to $Q = 4.7 \pm 0.5$ MeV. The intensity of this proton group was about one thirtieth of that of the group of Q-value 3.3 MeV and one thousandth of the total number of protons emitted. According to POLLARD (33) the 0.66 MeV Q-value belongs to the B^{11} (αp) C^{14} reaction.

KANNE (34) investigated the discrepancies in the experiments of Pose and of

Chadwick and Constable regarding the protons emitted by aluminium under α-bombardment. He used a thick aluminium target, polonium α-particles and a vacuum tube electrometer to detect the protons. The structure of the groups observed agreed with the results of Chadwick and Constable, while the ranges of the groups were in better accordance with those found by Pose.

Maier-Leibnitz and Maurer (35) searched in vain for the 4.7 ± 0.5 MeV Q-value proton group found by Brubaker and Pollard. Before that, they had found a 91 cm proton group from boron bombarded with polonium α-particles. This gives $Q = 3.7$ MeV, which they suppose to agree with the calculated value for a transition to the ground state of C^{13}.

Joliot and Zlotowski (36) used a Wilson chamber with a magnetic field of 16 000 gauss for investigating the energy of the protons from boron bombarded with polonium α-particles. They found one proton group with $Q = 4.4$ MeV ($E_\alpha = 4.8$ MeV). The next Q-value was 2.0 MeV. Joliot and Zlotowski thought the earlier Q-value 3.1 MeV, found by other investigators, could be the mean value of these two groups.

Blau (37) and Wambacher (38) made the first experiments on recording protons from (αp)-reactions by means of photographic emulsions. Merhaut (39) bombarded an aluminium layer of 1.3 mm air equivalent with polonium α-particles, the energy of which was varied by aluminium absorbers. The protons were recorded at right angle ($\vartheta = 90^\circ \pm 3^\circ$) by photographic emulsions and the ranges in the emulsion measured. He found four proton groups. The Q-value for the three largest groups were 2.25, 0.01 and -1.15 MeV.

Jentschke (40) using a coincidence ionization chamber thought he found the long range proton group from boron bombarded with Po-α. Jurka used the photographic method, developed by Merhaut (41). The highest Q-value he found was 3.85 MeV.

Benson (42) bombarded aluminium with 7.3 MeV α-particles from a cyclotron and studied the p, γ-coincidences. No γ-rays accompanied the end group of protons. Thus, this group probably belongs to a disintegration to the ground state of Si^{30}. Most of the protons in the fourth group are not associated with γ-rays and are therefore knock-on protons. Benson found the Q-values 2.22, -0.06, -1.44, and -2.20 to -2.63 MeV. The geometry was, however, poor.

Brolley, Sampson and Mitchell (43) bombarded aluminium with 22 MeV cyclotron α-particles. The Q-values found were -1.27, -3.22, -4.96, -5.98, -7.04, -7.65 and -8.64 MeV.

Creagan (44) bombarded boron with 7.4 MeV alphas produced in a cyclotron and observed the maximum energy protons at right angle. He found $Q = 3.9$ MeV, which is within experimental error of checking mass spectrograph data. At 0° maximum energy protons correspond to a lower Q-value. There appears to be a strong asymmetry in yield of protons from the group. Other Q-values observed were 0.8, -0.5 and -1.1 MeV.

Roy (45) utilized a cloud chamber and studied chiefly the angular distribution of the protons from boron bombarded with Po-α. The Q-values found were 3.1, 0.42 and 0.09 MeV. Owing to the few protons observed there was no indication of the longest range group of protons.

Creagan (46) continued the measurements quoted above and used targets of natural boron and targets of 96 percent B^{10}. The α-particle energy was 7.45 and 6.64 MeV. Seven charged particle groups were observed. The Q-values

Table I

B $(\alpha\, p)$ C.

Author	Year	Ref. No.	Method	α-source	Q-values (MeV)						
Bothe, Fränz	1927, 28	11 12	Point counter	Po			(−1.4)*				
Chadwick, Constable, Pollard	1931	19	Ionization chamber	»					0.2		3.2
Heidenreich	1933	23	» »	»		−1.92				1.27	7.63
Paton	1934	24	Proportional counter	ThC′		−1.86	−0.78		0.35	3.1	
v. Baeyer	1935	25	p,γ-coinc.	Po							
Miller, Duncanson, May	1934	31	Geiger ball counter	RaC′			−1.0	−0.1	0.4	3.1	
Livingston, Bethe	1937	2	Report, corrections	»			−0.8	0.1	0.5	3.3	
Brubaker, Pollard	1937	32	Ionization chamber	»	**						4.7
Maier-Leibnitz, Maurer	1937	35	» »	Po	**					3.7	
Joliot, Zlotowski	1938	36	Wilson chamber	»	**			2.0			4.4
Jentschke	1940	40	Ionization chamber	»	**						
Jurka	1940	41	Photographic plate	»	**					3.85	
Creagan I	1949	44	Proportional counter	cyclotron			−1.1	−0.5	0.8	3.9	
Creagan II	1949	46	» »	»	−1.76 (recoil)	−1.57 (weak)	−0.31	0.07	0.31	0.85	4.07
Roy	1949	45	Cloud chamber	Po				0.09	0.42	3.1	

* Computed by Chadwick, Constable, Pollard, Ref. 19.

** Search only for the longest proton group.

Table II

Al $(\alpha\,p)$ Si.

Author	Year	Ref. No.	Method	α-source	*Q*-values (MeV)						
CHADWICK, CONSTABLE, POLLARD	1931	19	Ionization chamber	Po						−0.1	2.3
CHADWICK, CONSTABLE	1932	20	» »	»						~0.05	~2.3
DIEBNER, POSE	1932	21	» »	»						0.03	3.53
STEUDEL	1932	22	» »	»							
HAXEL	1933	26	Proportional counter	RaC, ThC			~−2.0		~0		~2.3
HAXEL	1934	27, 28	» »	» »			−2.6	−1.1	0		2.3
HAFSTAD	1933	29	Ionization chamber	Po							
DUNCANSON, MILLER	1934	30	Geiger ball counter	RaC′			−2.67	−1.53	−0.16		2.07
LIVINGSTON, BETHE	1937	2	Report, corrections	»			−2.49	−1.32	−0.02		2.26
STEPPAN	1935	39	Photographic plate	Po					0		2.2
KANNE	1937	34	Ionization chamber	»							
MERHAUT	1940	39	Photographic plate	»				−1.15	0.01		2.25
BENSON	1947	42	p,γ-coinc.	cyclotron	−(2.20→2.63)			−1.44	−0.06		2.22
BROLLEY, SAMPSON, MITCHELL	1949	43	Proportional counter	»	−8.64	−7.65	−7.04	−5.98	−4.96	−3.22	−1.27
LANDON	1950	47	Proton coincidence	»		−2.6		−1.3	0.0		2.4

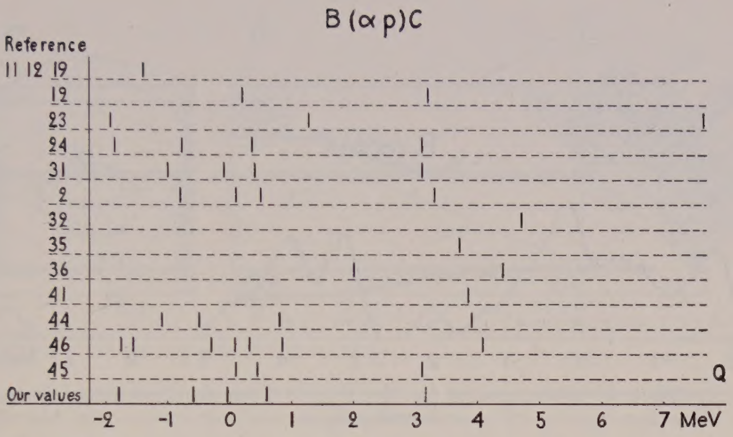

Fig. 1. Earlier Q-values for the B $(\alpha\, p)$ C-reaction, diagrammatically.

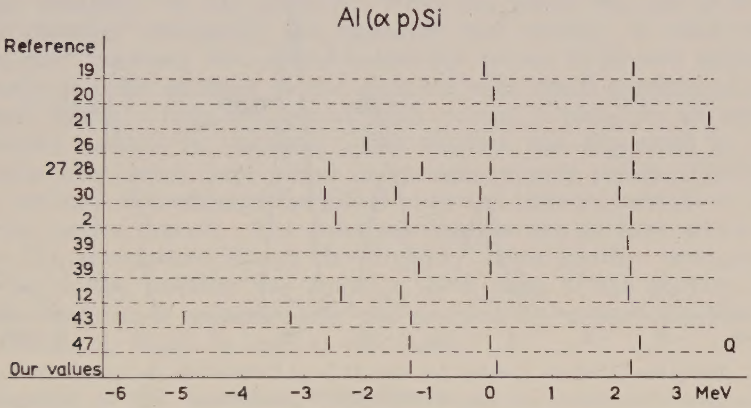

Fig. 2. Earlier Q-values for the Al $(\alpha\, p)$ Si-reaction, diagrammatically.

obtained were assigned as follows: $Q_1 = 4.07$ MeV to the B^{10} $(\alpha\, p)$ C^{13} reaction, $Q_2 = 0.85$ MeV to the B^{11} $(\alpha\, p)$ C^{14} reaction, $Q_3 = 0.31$ and $Q_4 = 0.07$ MeV to B^{10}. The next group gives $Q_5 = -0.31$ MeV if it is assumed, it is due to protons. If the group belongs to deuterons from the reaction B^{10} $(\alpha\, d)$ C^{12}, the Q-value equals 1.55 MeV in good agreement with the value 1.45 MeV predicted by mass differences. $Q_6 = -1.57$ MeV belongs to a very weak group observed only at 0°. Assignment is uncertain. $Q_7 = -1.76$ MeV is apparently due to recoil protons observed at 0°. Proton-gamma-coincidence observations were made for group assignments. LANDON (47) studied the Al^{27} $(\alpha\, p)$ Si^{30}-reaction by means of a proton-gamma-ray coincidence technique. The protons were observed at right angle. The Q-values found were 2.4, 0.0, -1.3, -2.6 ± 0.2 MeV.

The results previously obtained are summarized for convenience in Tables I and II. The Q-values found are marked in Figs. 1 and 2. The degree of agreement is very clearly seen from Figs. 3 and 4. The curves are obtained in the following manner. A triangle, the shape of which appears for instance for the

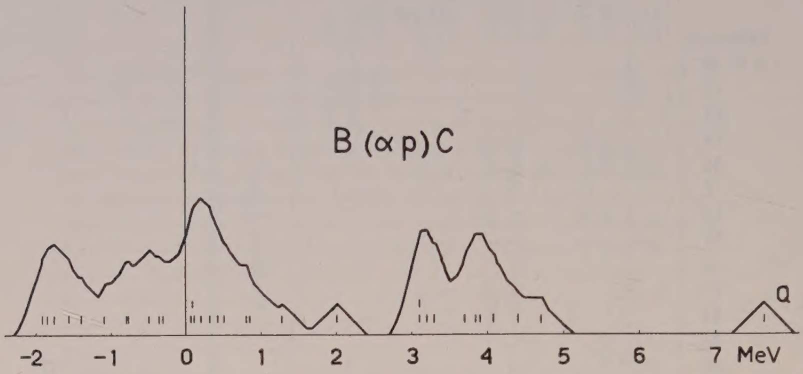

Fig. 3. Probable Q-values (peaks) for the B (α p) C-reaction, computed from earlier investigations.

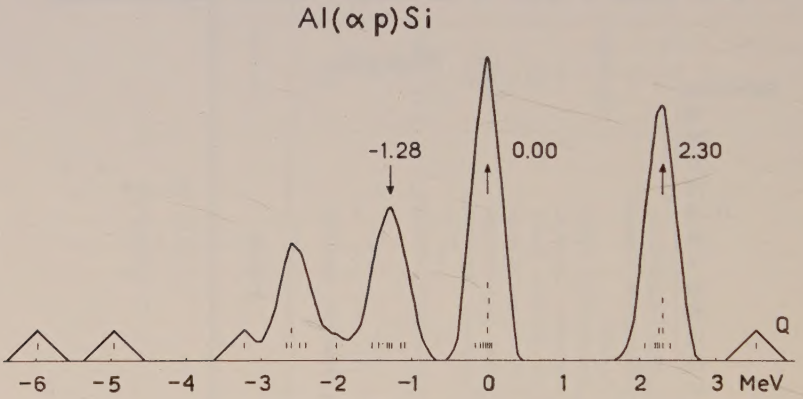

Fig. 4. Probable Q-values (peaks) for the Al (α p) C-reaction, computed from earlier investigations.

Q-value -4.96 in Fig. 4, is assigned to each Q-value found by earlier authors. These triangles mean, that the Q-values regarded may be wrong by ± 0.4 MeV and that the probability for the correctness of the Q-value Q_1 instead of the experimental value Q_0 is proportional to $|Q_0 - Q_1|$. When all these diagrammatical probabilities are summarized, we get the curves drawn. It is immediately seen, that the Q-values for the Al (α p) Si reaction are much better known than those for the B (α p) C process. A new examination of the latter Q-values seems therefore to be very desirable.

In this paper we have investigated the Q-values for both reactions. The Al-reaction was used in the main as a check for the new method, which is described in the next paragraph.

3. Experimental arrangement. Fig. 5 shows two sections of the apparatus employed. E is the photographic plate, T the target and Po the polonium sample. S_E and S_T are springs, which keep the plate and the target in position. The polonium sample was deposited on a nickel plate. The diameter of the

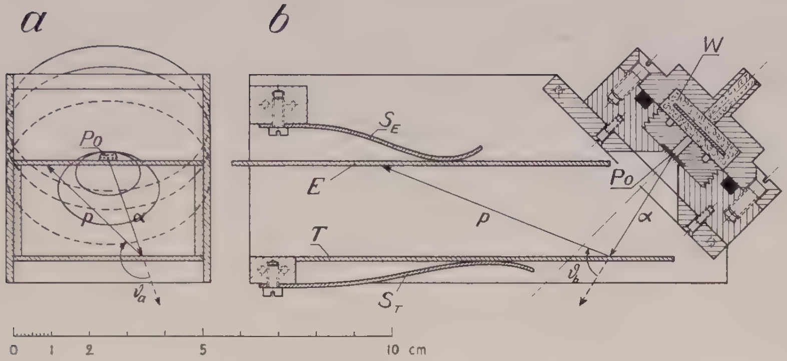

Fig. 5. Experimental arrangement for investigating proton tracks from different targets T, bombarded with α-particles from a polonium source Po. E=photographic plate.

active spot was six mm. No direct α-particles from the polonium source could reach the emulsion on the plate E. When the frame was put in a chamber and this chamber evacuated, the exposure was started. A mica window of 0.67 mgs./cm^2 thickness was placed before the source to prevent contamination of the walls and the surfaces in the chamber with small amounts of polonium. (As is well known a large number of small clusters of polonium are spread from an uncovered sample in vacuum). The window was supported by a lattice. The sample was always kept either in vacuum or in a carbon dioxide atmosphere to prevent oxidization of the nickel plate and thus an increase of the thickness of the active deposit. The polonium sample was at first always kept in the same CO_2-atmosphere behind the window. Later another arrangement was used, in which the pressure was equal on both sides of the window. By evacuating the chamber a contamination was avoided by means of cotton wadding (W) in a channel, which is seen in the figure.

4. Scanning of the emulsions. The emulsions were scanned with a binocular "Ortholux" research microscope. Generally a magnification of 300 was used. The microscope is equipped with a mechanical stage designed so that measurements may be made over slides measuring 100 mm × 50 mm. The vertical dimension is measured by the fine focusing mechanism, which is graduated to 1 μ.

The length of the tracks is measured by a screw micrometer and the direction of the track by means of a graduated circle. As a rule the scanning of the emulsions was performed with a dark ground condenser.

5. The polonium sample and the energy distribution of α-particles. It was already mentioned that the polonium was deposited as a 6 mm diameter spot on a nickel plate. The sample was purchased from the Canadian Radium & Uranium Corporation[1], New York, and had a strength of about 25 mC in February 1950.

The polonium sample was mounted in the brass holder with the mica window

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[1] The purchase was effected by the firm Erik Lindblom & Co., Sthlm.

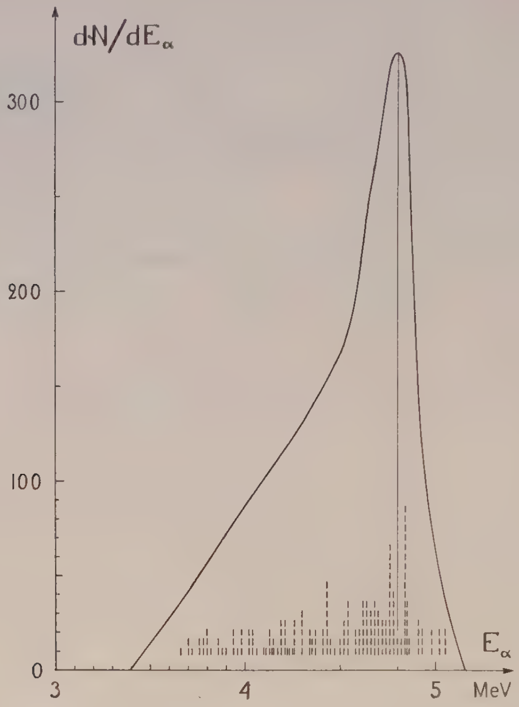

Fig. 6. Energy distribution of the α-particles from the polonium source behind the mica window.

and the assembly (without the photographic plate and target holder described above) was placed in a brass tube of 50 cm length close to one end of the tube. A plate holder in the other end of the tube held an Ilford Nuclear Research plate type C 2 of 100 μ emulsion thickness in such a position that the angle between the paths of the α-particles and the surface of the emulsion was about 5°. The tube was then evacuated. The mica window was perpendicular to the axis of the tube.

The length of the tracks was measured and the energy of the α-particles computed from the range-energy relation by LATTES, FOWLER and CUER (48). Fig. 6 shows the energy distribution found for the α-particles. The most frequent energy value is 4.80 MeV, and the half width of the distribution is 0.40 MeV.

6. Calculation of the *Q*-values. Fig. 7 shows diagrammatically the target, the polonium sample and the emulsion, the thickness of which is greatly exaggerated for the sake of clarity. a is the length of the path of the α-particle from the source Po to the disintegration site. The coordinates for this point are u and z. p is the length of the proton path and l is the length of the proton track in the emulsion. The coordinates for the starting point S of the track are ξ and η, and d is the distance of S from Po. The following formulas are easily obtained from the figure (all distances in mm.)

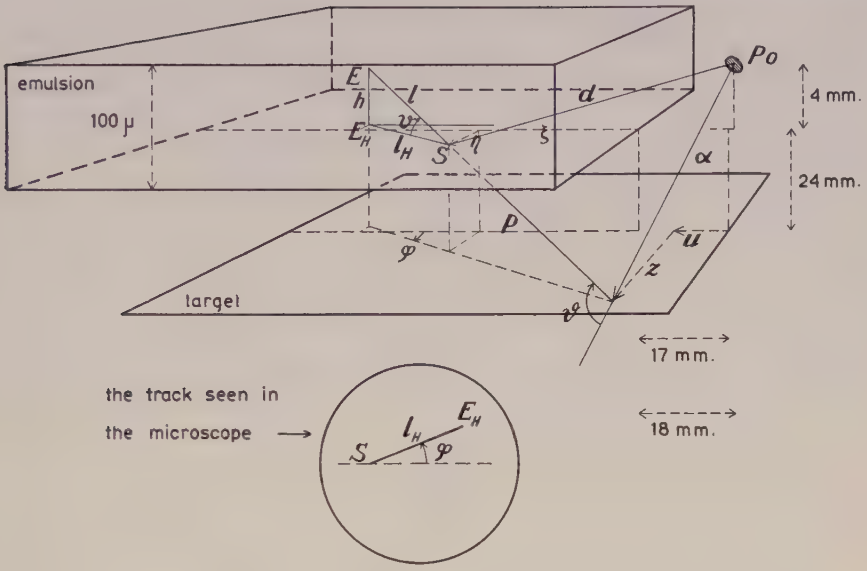

Fig. 7. The experimental arrangement with notations for the variables.

$$
\begin{aligned}
v &= \arctan h/l_H \\
p &= 24/\sin v \\
z &= \eta + 24 \cot v \cdot \sin \varphi \\
u &= \xi + 17 - 24 \cot v \cdot \cos \varphi \\
a^2 &= (u+1)^2 + z^2 + 28^2 \\
d^2 &= (\xi + 18)^2 + \eta^2 + 4^2 \\
\cos \vartheta &= (d^2 - p^2 - a^2)/2\,a\,p.
\end{aligned}
\tag{3}
$$

The Q-values are then found from the formula (2). h is obtained when the difference between the readings on the fine focusing screw corresponding to S and E is multiplied with the calibration factor f (about 3.8). This in turn is found when the surfaces of the emulsion layer are focused and the corresponding reading difference is compared with 100 μ.

Instead of computing the $\cos \vartheta$-values by means of the formulas (3), the $\cos \vartheta$-values are easily calculated by simple geometrical constructions. Fig. 8 shows an example corresponding to the case in Fig. 7.

7. Correction for oblique passage through the window. The most frequent energy for the α-particles is 4.8 MeV only for particles, which pass perpendicularly through the window. The original energy of the α-particles, 5.3 MeV, is diminished because of the thickness of the sample itself and the thickness of the mica window. The decrease in energy 0.5 MeV, corresponds to a certain decrease Δr in range. If the angle between the path of the α-particle and

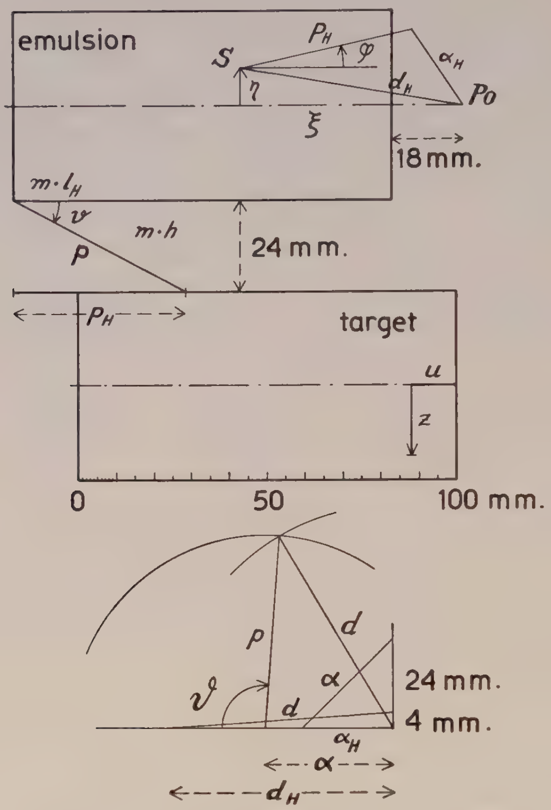

Fig. 8. Geometrical construction of the angle ϑ.

the window normal is w, the decrease in range will be $\Delta r/\cos w$ and, with sufficient accuracy for correction purposes, the decrease in energy 0.5 MeV/cos w. This correction was made for each α-particle energy E_α when the Q-values were computed. The simplest method for this computation is to draw

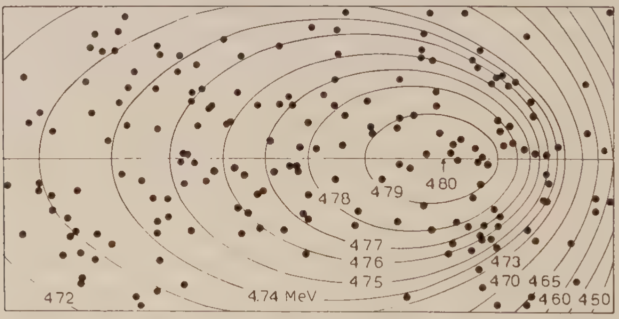

Fig. 9. Map of the boron target. The dark circles mark the places where disintegrations have occurred. The ellipses give the corresponding α-particle energy.

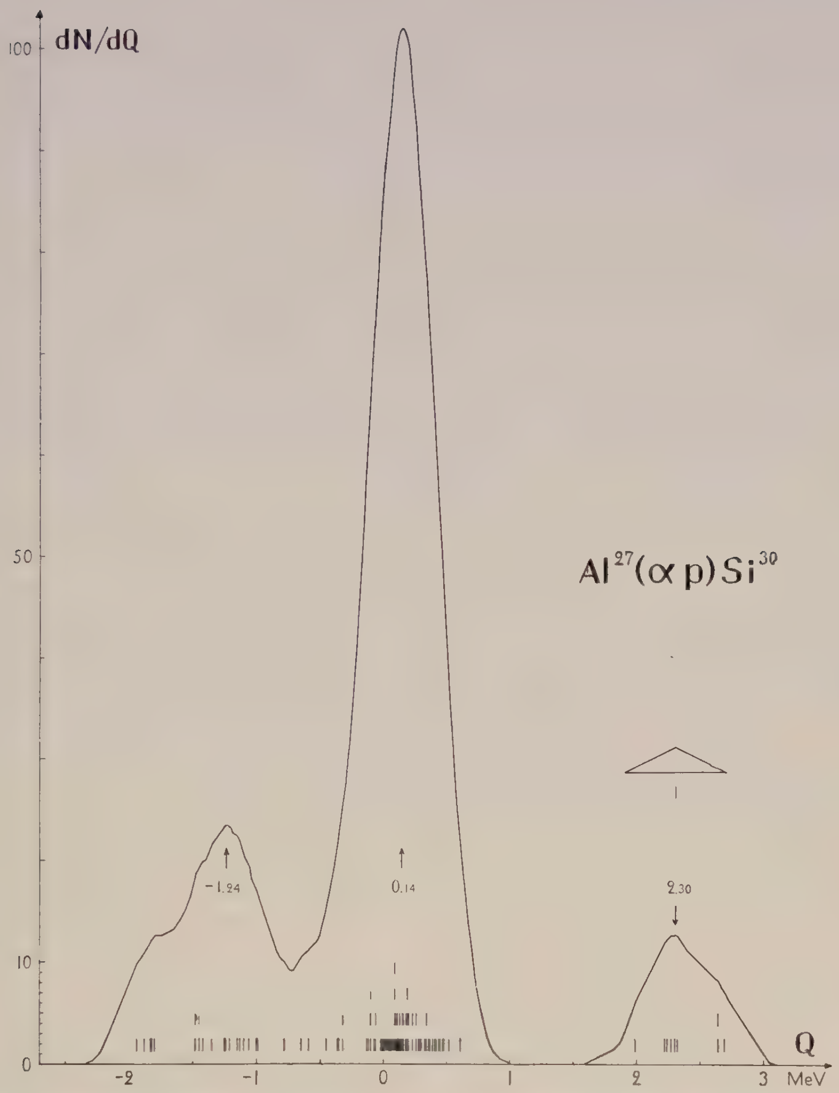

Fig. 10. The most probable Q-values for the Al $(\alpha\ p)$ Si-reaction, computed from the Q-values obtained from 100 proton tracks.

the curves for $E_a = 4.8$ MeV (this curve degenerates to a point), $E_a = 4.79$ MeV, 4.78 MeV etc. on a map of the target. These curves are ellipses and are easily constructed. When the coordinates u und z are computed, the corresponding E_a-value is immediately obtained from the curves. Fig. 9 shows such a map of the boron target with the ellipses for constant α-particle energies and all the places (dark circles), where disintegrations have been observed.

8. Bombardment of aluminium. The aluminium target consisted of a 0.15 mgs./cm^2 foil over a brass plate. An Ilford Nuclear Research plate type C 2

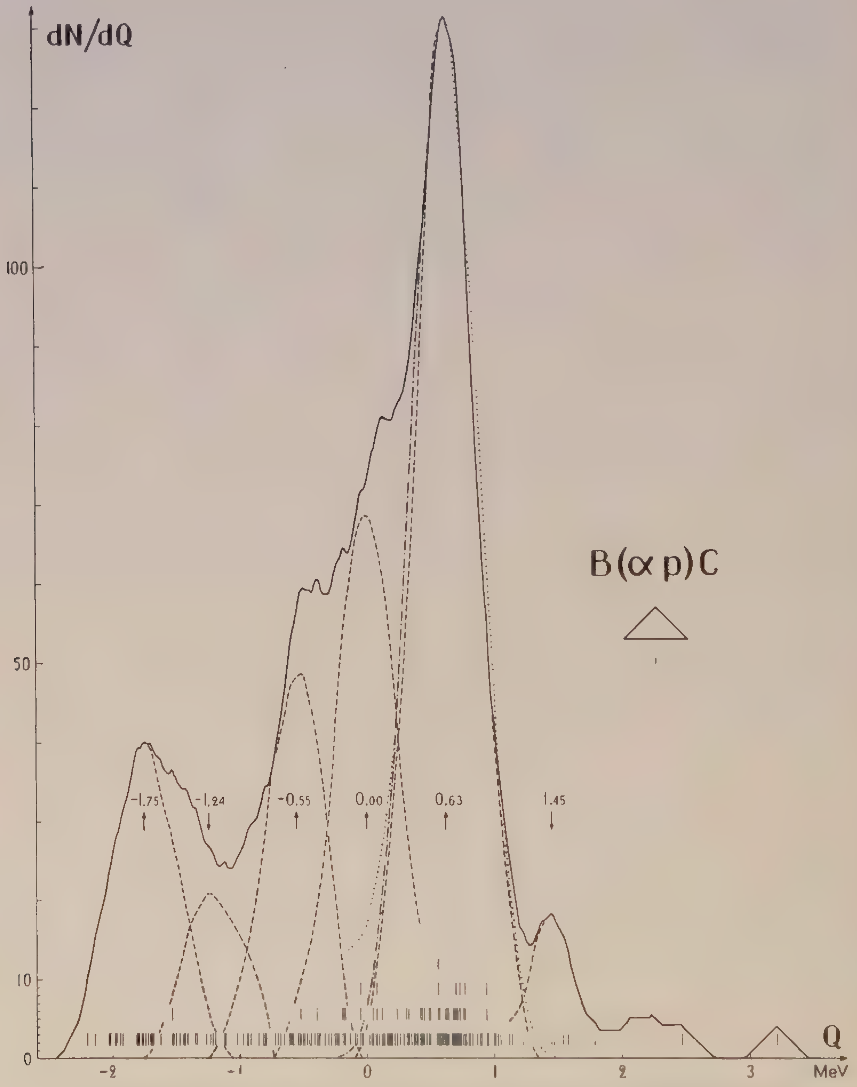

Fig. 11. The most probable Q-values for the B (α p) C-reaction, computed from the Q-values obtained from 200 proton tracks.

of 100 μ thickness was exposed to protons from the aluminium target for about 21.5 hours (June 10th, 1950). The plate was developed according to the description accompanying the plates. The Q-values obtained are presented diagrammatically in Fig. 10. To each one of these 100 Q-values is assigned a triangle of a shape which is seen to the right in the figure. The triangle means that any Q-value might be wrong with ± 0.4 MeV, depending on the energy distribution of the α-particles, and that the probability for the correctness of the Q-value Q_1 instead of the experimental value Q_0 is proportinal to $|Q_0 - Q_1$.

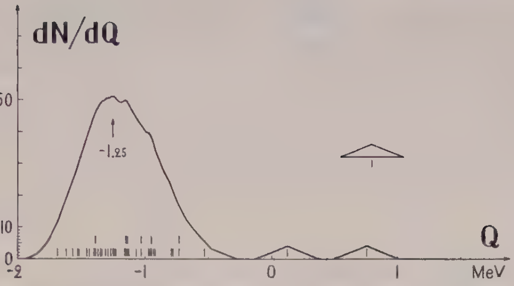

Fig. 12. The false Q-value, obtained from recoil protons from the mica window and elastically scattered by the iron target backing.

The curves are obtained when all these diagrammatical probabilities are summarized.

The figure shows that there are three groups of Q-values, *viz.* — 1.24, 0.14 and 2.30 MeV.

The agreement between these values and those of the more recent investigations by other authors is very good, as is seen from Table II and from Fig. 2. Fig. 4 gives for the most probable mean of the corresponding earlier Q-values

— 1.28, 0.00, and 2.30 MeV.

9. Bombardment of boron. The boron target consisted of boron of thickness 0.048 and 0.056 mgs./cm^2, respectively, evaporated on two iron plates.[1] Exposure was made for 16.3 hours (May 27th, 1950). The distribution of 200 Q-values is given in Fig. 11.

For the sake of comparison the aluminium 0.14-group is placed on the main Q-group for boron. The aluminium comparison line is marked with points. The agreement between the lines is remarkably good. It is also seen that the aluminium line is quite symmetrical. This is seen when the right side of the line is reflected in a vertical through the top of the line. This reflected image is marked with point-lines. This line and the point-marked line differ only at the base of the line.

The very symmetrical character of the aluminium Q-line gives reason for an attempt to resolve the Q-spectrum for the B $(\alpha\, p)$ C-reactions. The result is seen from the figure. There are five principle groups, with the Q-values — 1.75, — 1.24, — 0.55, 0,00, and 0.63 MeV, respectively. The group at 1.45 MeV is uncertain. The group — 1.24 MeV is due to recoil protons from the mica window, when these protons are elastically scattered by the iron atoms. This was proved by the following experiment. The boron targets were replaced by iron. Fig. 12 shows the distribution of the Q-values obtained from the 75 proton tracks investigated. The top value — 1.25 MeV is in very good agreement with the value — 1.24 MeV found by the procedure described above.

Hence, we have found the following four Q-values for the B $(\alpha\, p)$ C-reaction

— 1.75, — 0.55, 0.00, and 0.63 MeV.

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[1] We are indebted to Ing. J. E. FLINTA at the laboratory of Dr. S. EKLUND, Försvarets Forskningsanstalt, Stockholm, for the courtesy of putting the boron targets at our disposal.

10. Comparison between Creagan's and our Q-values for the boron reaction. The most reliable earlier investigation seems to be the second one of Creagan. He found the Q-values

$$-1.57, \quad -0.31, \quad 0.07, \quad 0.31, \quad \text{and } 0.85 \text{ MeV}.$$

If these values are diminished with 0.21 MeV, we get

$$-1.78, \quad -0.52, \quad -0.14, \quad 0.10, \quad \text{and } 0.64 \text{ MeV}.$$

These Q-values agree quite well with our values if we consider our 0.00 MeV group to be the unresolved mean of his -0.14 and 0.10 MeV groups. For clearness these lower Creagan values are compared with our Q-values in Fig. 13.

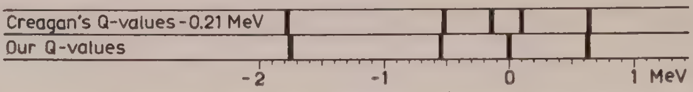

Fig. 13. Comparison between ours and Creagan's Q-values, the latter values reduced by 0.21 MeV.

Now the question arises whether Creagan's Q-values are too high or our values are too low. The good agreement with our Q-values and those of earlier investigators for the Al $(\alpha\, p)$ Si-reaction rather sharply supports the correctness of our values in the case of B.[1] Furthermore, a careful examination of the figures reproduced in Creagan's paper seems to indicate that the manner, in which he has determined the proton energy, is not free from objections. He counted largest proton pulses, and got the energy by taking the extrapolated value of the steepest part of the counts *versus* energy. Thus the energy was obtained as the point of intersection between this steepest part and the energy axis, even if there was a large background from other lines in the neighbourhood. We take for instance the line marked 6 in Fig. 4 of Creagan's paper. First, this line should obviously be marked 5, because the line 6 "is very weak and was observed only at 0°". We have computed the Q-value from the position marked 6 and get -0.42 MeV (this should correspond to Creagan's value -0.31 MeV). On the other hand, if we take the lowest point of the curve, that is the point of intersection of the steepest part of the curve with the increasing line 4, we get the Q-value -0.67 MeV. This value is 0.25 MeV lower, i. e. about equal to the difference between Creagan's and our Q-values as stated above.

11. Accuracy of our method. With a particular knowledge of the α-particle energy and of the elements ξ, η, φ, l_H, h for an $(\alpha\, p)$-reaction one single proton track should be sufficient for determining one of the exact Q-values. The curvature of the tracks and the energy spread of the α-particles make, however, the investigation of many tracks necessary.

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[1] Note added in proof; J. L. PERKIN (Phys. Rev. *79*, 175, 1950) found the Q-values 4.08 ± 0.12, 3.35 ± 0.25, 0.65 ± 0.15, 0.15 ± 0.15 and -0.57 ± 0.15 MeV with the relative yields for the first four groups 10 : 1 : 100 : 50. Our single proton track with $Q = 3.21$ MeV (Fig. 11) and our -0.55, 0.00, and 0.63-MeV groups are in good agreement with Perkin's result.

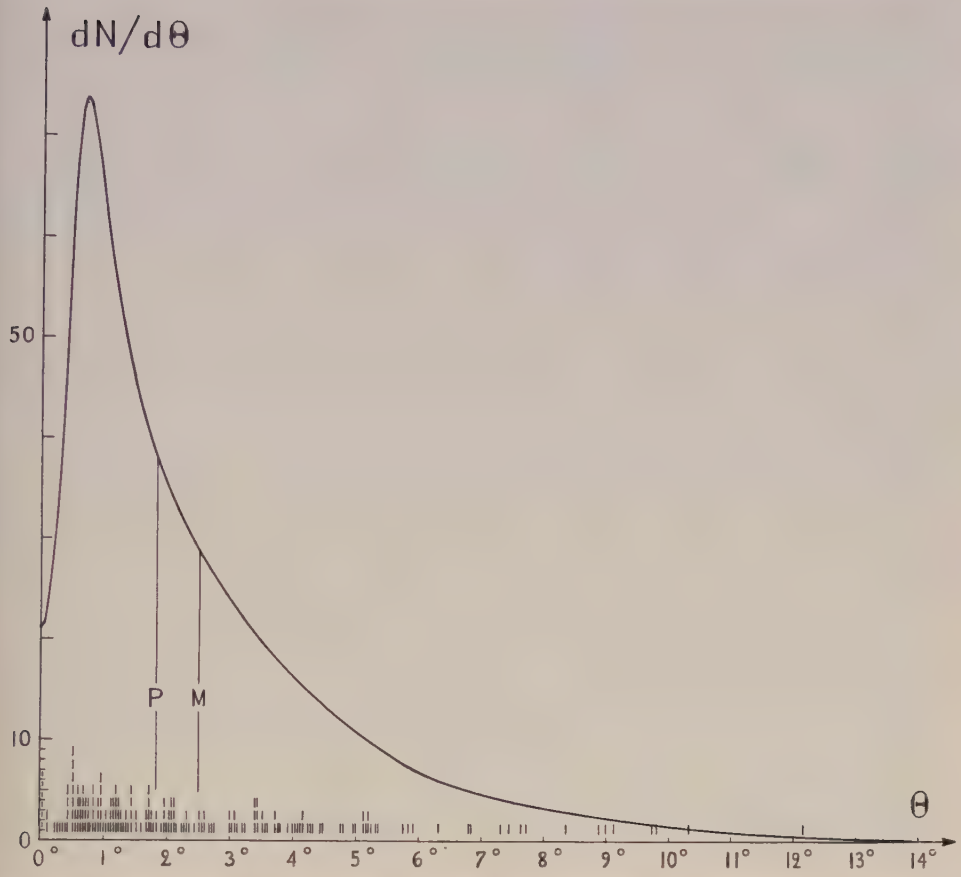

Fig. 14. Distribution of horizontal projection of angle difference, multiplied by cos v, between tangent to proton track at starting point and the chord between starting and end-point of the proton.

The question now arises, how large is the influence of the scattering of the protons. We have made measurements of the horizontal projection of the angle difference between the tangent to the proton track at the starting point and the chord between starting and end points. This difference, multiplied with cos v, may be called θ. Fig. 14 shows the distribution of tracks versus θ. Half of the number of tracks have a deviation smaller than $1^\circ.8$ and the mean value is $2^\circ.5$.

The mean error in measuring φ is $0^\circ.36$ and the mean error in measuring θ is $0^\circ.4$.

The largest errors arise from the measuring of h. It is difficult to focus the end of a track, and physiological properties of the eyes may cause different results when h is measured again after some time interval or by some other person. There is e. g. a constant difference in the measurements by two of us. The depths h are almost 2 μ greater when measured by E. H. than by

H. S. If this 2 μ is regarded as the size of the error in the depth measurements, it is seen that the error in the angle v caused by error in the depth measurement will be much larger for short tracks than for longer. The shortest tracks belong to $Q = -1.79$ MeV. For large ξ-values and small u-values we get $v \approx 14^\circ$, $E_p \approx 1.5$ MeV, $l = 26\ \mu$, and thus an error in the angle of about 4°. This circumstance causes the largest change in ϑ, which is altered from e. g. 90° to 58°. Instead of the correct value -1.79 MeV, we then get -2.23 MeV. In any case, this value may be of interest, if we have many such values, because we then get a distribution, from which the most probable Q-value can be computed. The spread in Q is of the same order of magnitude as the spread in energy of the incident α-particles. There is no reason to use only good tracks before we have a more monoenergetic α-source. It is easily shown by the formula (2), that the spread in Q caused by the spread in α-particle energy is of the same order of magnitude as this latter spread. In fact a little smaller, because of the factor $\left(1 - \frac{M_\alpha}{M_r}\right) = \frac{9}{13}$.

We have chosen the most disadvantageous example. The personal error could, of course, partly be corrected for. However, if we have a more monoenergetic α-particle source, we can exclude those tracks, for which the change in ϑ is large. The best tracks are obviously those, for which $z \approx \frac{1}{2}\eta$ and $u + 1 \approx \frac{1}{2}(\xi + 17)$. A moderate error in v or φ will then be without influence on ϑ.

In the present case the half width in the energy distribution of the α-particles is about 0.4 MeV. This will cause an uncertainty in a boron Q-value, computed from one single track, of about ± 0.3 MeV. The uncertainty in a Q-value, computed from ten tracks, will then be about ± 0.1 MeV and so on. However, if there are sharp resonance groups, differences may occur, because we do not know the energy of the α-particles, which causes the proton group. This energy may differ from the peak value 4.80 MeV.

Our investigations on the $(\alpha\, p)$-processes by means of the present method are being continued.

SUMMARY

1. A survey is given over earlier investigations concerning $(\alpha\, p)$-reactions on aluminum and boron. Previous Q-values are presented in Table I and II and in Figs 1–4.

2. Earlier Q-values for the B $(\alpha\, p)$ C-reaction are inconsistent and require new investigations.

3. A new arrangement for measuring Q-values by means of modern photographic nuclear research emulsions and a polonium α-particle source is developed and described.

4. The Q-values obtained for the Al $(\alpha\, p)$ Si-reactions, viz. -1.24, 0.14 and 2.30 MeV agree very well with those given by recent investigators.

5. The Q-values found for the B $(\alpha\, p)$ C-reactions were -1.75, -0.55, 0.00, and 0.63 MeV. The value 0.00 MeV may, however, be a mean of two or more unresolved Q-values.

6. The result for boron is compared with those of Creagan and Perkin.
7. The accuracy of the new method is discussed.

Nobel Institute of Physics, Academy of Sciences, Stockholm.

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Uppsala 1951. Almqvist & Wiksells Boktryckeri AB

Table 2

Source of radiation	Millirem[1] per week	
	Min.	Max.
External sources: Cosmic rays	0.6 (sea level)	0.7 (500 m above sea)
γ rays	0.3 (in nature)	9 (in houses)
Internal sources:		
K 40 content in skeleton 0.06 % K	0.1	
muscles 0.37 " "	0.7	
whole body 0.20 " "	0.4	
Ra in skeleton (50 % Rn assumed to escape from skeleton)		
In cremation ashes: acc. to KREBS	<6	250
acc. to HURSH & GATES	0.25	2.5
Total Ra-content acc. to the author's prel. measurements (25 % Rn ass. to escape from body)	—	100

It is difficult to judge whether there was an actual difference in the radium content of the subjects tested or whether, as HURSH and GATES found probable, KREBS' higher figures may be ascribed to impurities originating from the cremation method.

It is, however, of great value to know what is the magnitude of the normal radium content. If KREBS' average values are correct, the radium must be the main origin of the natural ionizing radiation, at least in the skeleton. If, on the contrary, the value of HURSH and GATES is accepted, the radium content contributes only slightly to this radiation and the γ-radiation from the surroundings plays a relatively more important rôle. This is obvious from Table 2, in which the α-radiation is assumed to have about 10 times higher biological efficiency than the β- and γ-radiation.

From the foregoing it is apparent that investigations with a method for testing living persons with regard to their radium content may be of interest. Such a method has been used by, for instance, HESS and MCNIFF[2] for measuring radium contents of the order of 0.1 ± 0.01 μg Ra.

The apparatus described in the present paper[3] was designed mainly for investigations of persons engaged in work with long-lived γ-radiating substances, principally radium and thorium. Although it has not proved sufficiently sensitive to solve the forementioned question definitely, the experience gained shows that the method is promising. With some improvements it should allow the determination of radium quantities down to amounts of a magnitude below which biological effects of the radium content may probably be disregarded in comparison with those of the other sources of ionizing radiation. The method may be used to advantage together with determinations of radon in the breath.

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[1] Milliroentgen equivalent man (roentgen unit, r, corrected for biological effect).

[2] V. F. HESS and W. T. MCNIFF, Am. Journ. Roentg., 57, 91, 1947.

[3] A report on the apparatus has been given at the meeting of the Scandinavian Society for Medical Radiology in June 1949 and at the 6th Intern. Congress of Radiology, July 1950.

Method and apparatus

The principle of the method is seen from the wiring diagram in Fig. 1. The pressure ion chamber A (25 kg/cm^2, N_2) is built up of 10 tubes, 22 cm in diameter and 125 cm in length, giving a total volume of about 400 l. The central electrodes have guard rings and are connected to the grid of an electrometer tube F and a 10^{12} ohm resistance B. The tubes are charged by means of a perspex-insulated dry battery of 900 V and covered with an earthed casing. They are arranged concentrically around a cylinder 50 cm in diameter, in which the subject to be tested is placed. A berth formed by an aluminium plate with a curvature of approximately 50 cm has been found to be surprisingly comfortable. The time constant for the measuring system amounts to about 5 minutes.

The plate current of the electrometer tube (VX41) is balanced by means of the arrangement H and, when the ionization changes, the out of balance current is recorded on a roll of photographic paper by means of an overdamped Ising[1] toroid galvanometer G_2. A Cambridge spot galvanometer G_1 is used for visual observations.

The operating data of the electrometer tube are very carefully controlled and the apparatus is run continuously for several months.

Because considerable time is required for stabilization, there is a special potentiometer and a contact arrangement D, which can be used when starting the recording. The potential on the earth side of the high resistance G, and the compensation current can be observed accurately (E and I) and are adjusted to a constant value 2–4 times a day. The atmospheric pressure is recorded with a microbarograph in order to be able to correct for slow changes in the cosmic radiation. The temperature of the apparatus is also recorded.

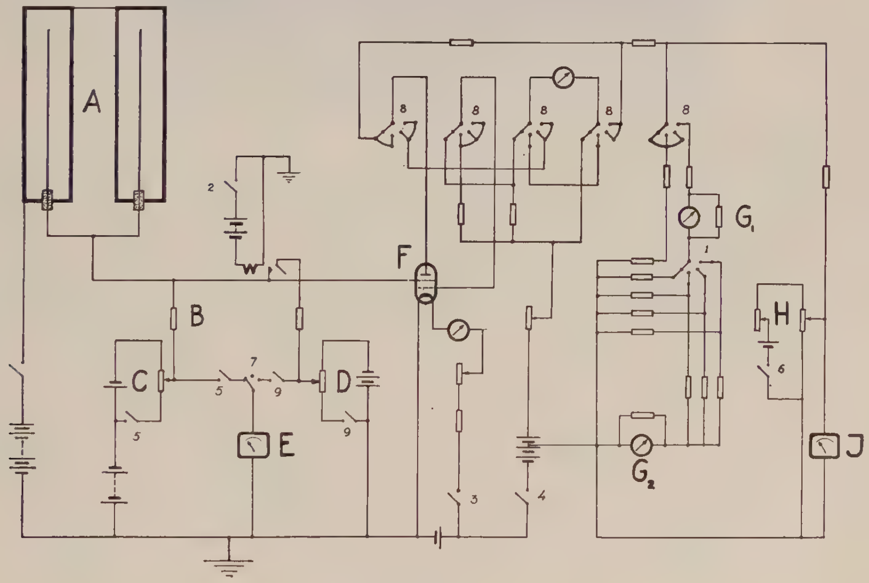

Fig. 1.

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[1] G. Ising, Ark. f. Mat., Astr. o. Fysik, 36 A, 1, 1948.

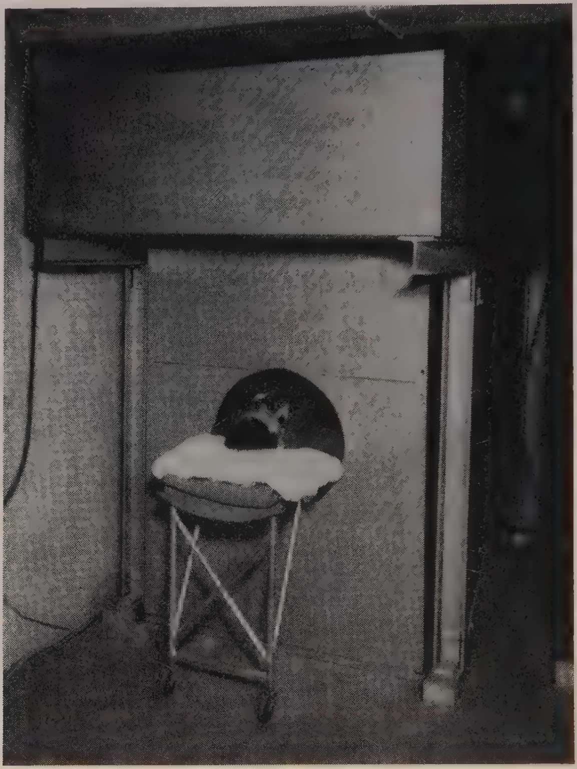

Fig. 2.

The apparatus (Figs. 2–4) is placed below the ground and shielded by concrete walls and water tanks, ensuring that the γ-radiation from the surroundings is comparatively constant. Hitherto, there has been insufficient space for effective water shielding. Occasionally, disturbances caused by radioactive preparations used in surrounding laboratories have also been noted.

The constancy of the sensitivity is checked every night by means of an automatically moving radium preparation of 0.1 μg radium which travels on a wire from its wall-position down to the centre of the ion chambers and remains there for 4 hours.

The absorption of the γ-radiation of the surroundings in the person to be tested is determined by means of rubber dummies of different volumes and the same relative cross-sections as average human bodies. They are filled with distilled water, of which the quantity can be varied from 50 to 75 kg. The dummies are also used for estimating the absorption of the γ-radiation from radioactive substances. Thus, measurements are made of the radiation from radium preparations in various positions inside the dummies and from the normal amount of K 40 (1.5 MeV γ-ray energy), by using solutions of different potassium salts.

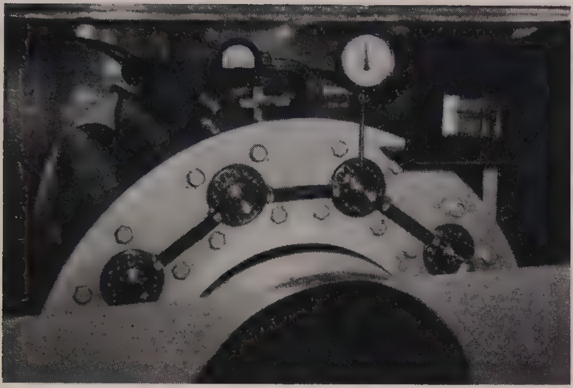

Fig. 3.

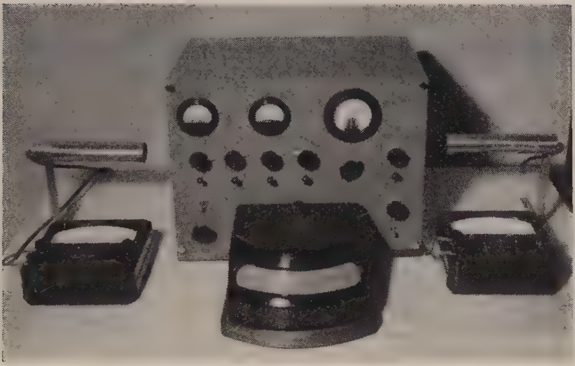

Fig. 4.

Preliminary results

Records are made with the radium preparation, the dummies and the persons to be tested, a zero-record being made between each of them. All these parts of the records are run for at least 2 hours. A typical record is shown in Fig. 5.

The variations which at present limit the accuracy of the method are caused by:

the statistical variations of γ-rays from the surrounding material and of cosmic rays;

the slow variations of the cosmic rays due to changes in atmospheric pressure and other circumstances and

the lack of sufficient shielding against γ-radiation from surrounding material. This necessitates a correction for body absorption of a magnitude corresponding to about 0.055 to 0.08 μg Ra in air for body weights of 50 to 80 kg.

To these sources of errors must be added the uncertainty of the correction for the absorption in the body of the γ-radiation to be measured. This is

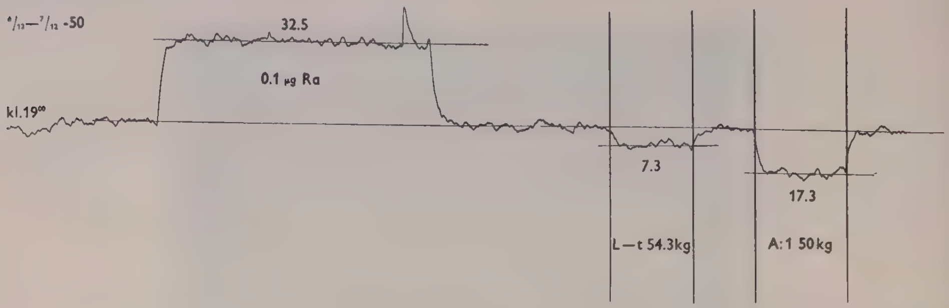

Fig. 5. A typical record showing a calibration deflection with 0.1 μg Ra and two absorption deflections, one with a person having a considerable radium content and one with a water-filled dummy.

difficult to determine on the grounds of the unknown distribution in the body of the radioactive element in question.

Some of the results obtained hitherto are assembled in Fig. 6, which shows the absorption values of the persons tested and of the rubber dummies, divided by the slightly differing values recorded with a standard preparation of 0.1 μg Ra (maximum deviation during about one year ± 5 %). The two lowest curves were obtained by actual measurements. The absorption curve representing the body without γ-radiation is based on more than 50 records made with 50, 60, 70 and 75 kg of water. The 60 kg value was obtained with two dummies of different size (filled to a different extent) giving surprisingly similar results. The curves for 0.01—0.20 μg Ra were calculated with the use of observations of the radiation from a radium preparation of 0.2 μg Ra in different positions in a dummy. These equidistant curves representing the radium content in the skeleton may have a maximum uncertainty of ± 15 % for the absolute values and are not corrected for excretion of radon and for K 40. The curve for K 40 (140 g K) was calculated from records with 50 and 70 kg of water containing about 3 kg of KNO_3 per 70 kg. The average content of potassium in the body was assumed to be 0.20 per cent.[1] Salts of different origin were tested. The present values must be regarded as provisional, since a purification of the salts for eliminating traces of radium will later be made. However, a test with an α scintillation counter gave negative results.

Under favourable conditions, the standard error can be reduced to ± 0.002 μg Ra eqv. with 6—8 observations for persons with no radioactive contamination. There may, however, be systematic errors of at least the same order of magnitude, caused mainly by the difference in absorption in the dummies and the experimental subjects and by the unknown distribution of the radium in the body.

The data for the persons tested are shown in Table 3 ("normal persons") and Table 4 (persons having been in contact with radioactive material). In the latter case precautions were taken to avoid the presence of surface contamination. Careful controls were therefore made with an α scintillation counter.

As is obvious from Fig. 7, there seems to be an increase in the probable radium content with increasing age. The results must, however, be regarded as com-

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[1] A. T. Shohl, Mineral Metabolism, Am. Chem. Soc. Monograph, Ser. 82, 1939.
L. Corsa Jr, J. M. Olney Jr, R. W. Steenburg, M. R. Ball and F. D. Moore, Journ. Clin. Invest. 29, 1280, 1950.

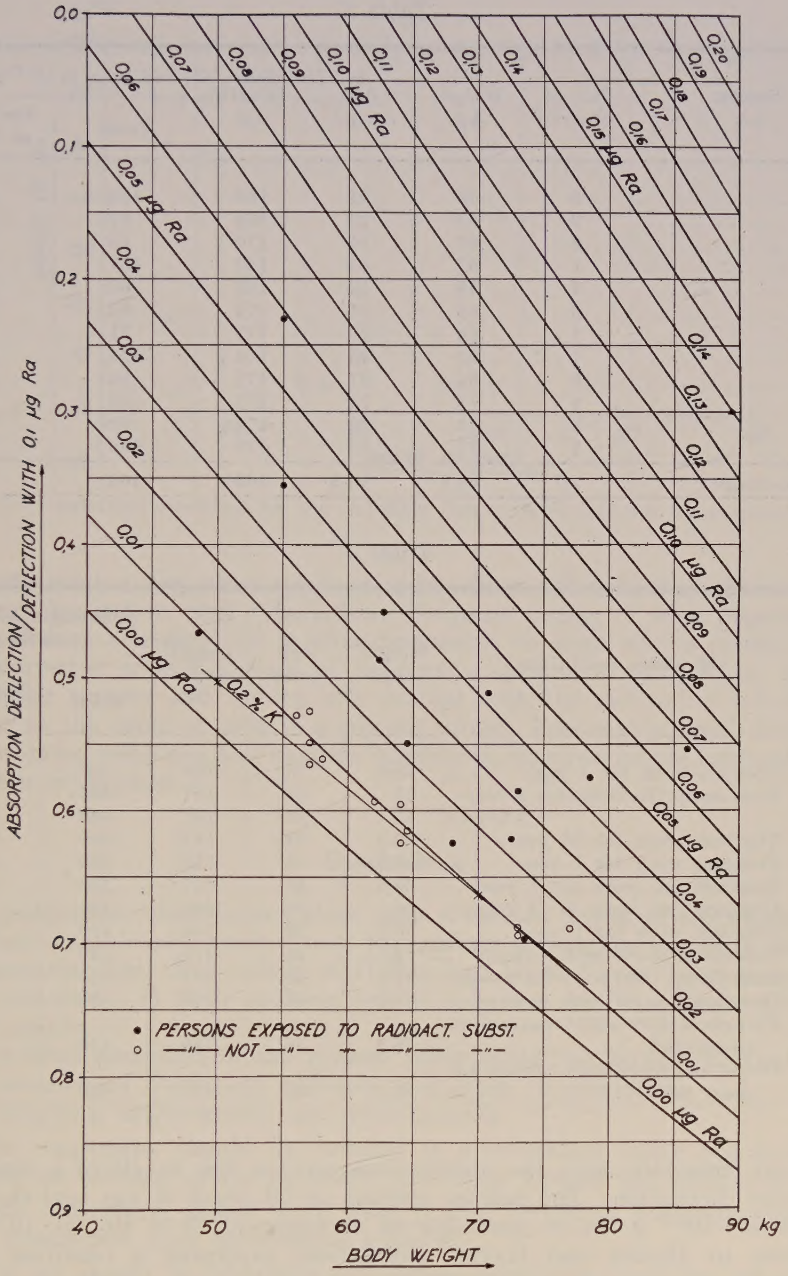

Fig. 6. The method of using the relative values of the absorption deflection is obvious from Fig. 5.

Table 3

Person no.	No. of observ.	Weight kg	Age years	Length cm	γ-radiation in 10^{-10} g Ra eqv.	
					Total	Per 70 kg of weight
n 1	3	56	31	165	114	142
n 2	2	57	23	164	67	82
n 3	2	57	15	170	93	114
n 4	1	57	43	157	137	168
n 5	7	58	19	163	90	109
n 6	5	62	25	173	92	104
n 7	1	64	27	171	71	78
n 8	1	64	48	164	124	136
n 9	8	64.5	27	171	96	104
n 10	2	73	25	175	86	83
n 11	5	73	23	174	97	93
n 12	7	77	58	176	165	150
Average:	3.7	63.6	30.3	168.6	103	114

Table 4

Person no.	Working conditions	Weight kg	Age years	Length cm	γ-radiation in 10^{-10} g Ra eqv.	
					Total	Per 70 kg of weight
1	Radium work for 1 year[1]	48.5	25	154	95	137
2	Lum. comp. (Ra) work for: 3 years	55	31	157	385	490
	3.6 years	55	31	157	590	750
3	Thorium work for 15 years	62.4	64	182	300	335
4	Thorium work for 1 year	62.7	36	159	370	415
5	Uranium ore work for 1 year	64.5	40	167	220	240
6	Uranium ore work for 3 years	68	36	166	140	145
7	Thorium work for 1 year	70.7	38	168	410	405
8	Occasional work with radium[1]	72.5	23	176	230	220
9	Lum. comp. (Ra) work for 8 years	73.4	32	171	310	295
10	Occasional work with radium[1]	73.5	32	174	90	86
11	Periodical work with large radium quantities[1]	78.5	36	180	450	400
12	Occasional work with radium for many years[1]	86.5	62	171	680	550

paratively uncertain since the observations are too few to afford a statistically significant correlation. The radium content at 50 years of age is of the magnitude of $60 \cdot 10^{-10}$ g Ra or, assuming an excretion of 25 % Rn, $80 \cdot 10^{-10}$ g Ra. According to Hursh and Gates' calculation, supposing a retention of 2 %, this would correspond to an average content in the water intake during life of $10–20 \cdot 10^{-15}$ g per ml which, for Swedish conditions, seems to be a reasonable value.

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[1] Open radium preparations.

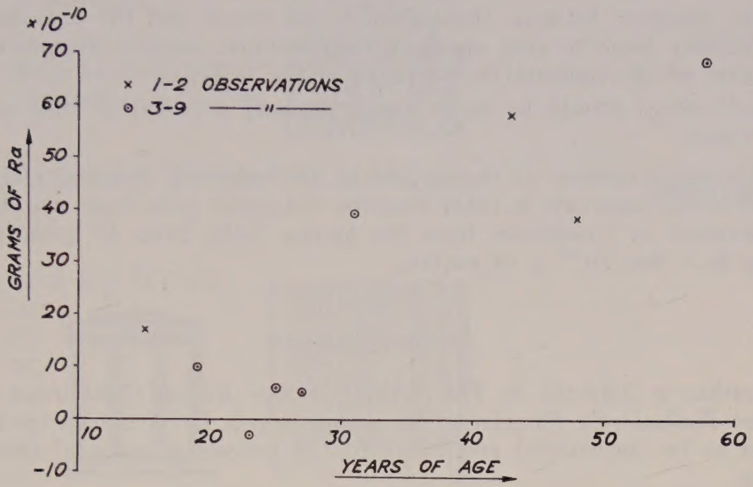

Fig. 7. γ-radiation corrected for the radiation due to K 40 but not for excreted Rn.

Already after a few years some persons working with radium or thorium show a marked increase in their γ-radiation. Frequent testing of such persons must be regarded as necessary. It is often impossible to state why a certain person has absorbed a greater amount of radioactive substances than others working in a similar manner and coming into contact with the radioactive substance in question to the same, or even to a greater, extent. Presumably, small differences in the working technique and hygienic conditions, which cause the contamination, have been overlooked.

Conclusions

The preliminary observations show with a certain degree of probability that, as Krebs[1] has already pointed out, the radium content in the body increases with increasing age. The values of Hursh and Gates[2] may be representative for the conditions in their subjects but do not seem to be in contradiction to Krebs' results.

The method described in the present paper could not be fully utilized owing to the presence of disturbing radiation and other circumstances.

The following improvements are recommended.

1. The apparatus should be installed in a laboratory below the surface of the earth, at a depth giving sufficient protection against the cosmic rays.

2. The laboratory should have walls (water) providing adequate protection against γ-radiation from surrounding material.

3. The air should be free from radioactive substances, and the temperature constant.

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[1] A. Krebs, Fundam. Radiol. 5, 89, 1939.

[2] J. B. Hursh and A. A. Gates, loc. cit.

4. The distance between the person to be tested and the chambers should be sufficiently large to give approximately uniform results, irrespective of the distribution of the radioactive substance in the body.

5. Each record should be made during as long a period of time as possible (e.g. 4 hours).

If all contact surfaces of the apparatus are correctly designed and the construction is also adequate in other respects, it appears possible to use the method for observation of γ-radiation from the human body down to quantities corresponding to a few 10^{-10} g of radium.

Acknowledgments

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